Fluctuations in confined nematic liquid crystals in a regime of critical wetting

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(6 December, 1998)

Within the macroscopic Landau-de Gennes approach, we examine the Gaussian normal mode fluctuations of semi-infinite nematic liquid crystals in a regime of critical wetting. It is argued that surface free-energy potentials which strongly suppress the long-range nematic order favor the appearance of bound biaxial nematic-director fluctuation modes, located in the domain occupied by the thermodynamic phase wetting the wall. Instead, substrates enhancing the orientational order promote the existence of uniaxial nematic-director local excitations. Close to the phase coexistence temperature both types of local exciations are strongly softened as compared to their bulk counterparts and acquire characteristic cusplike low-energy spectra. These spectrum peculiarities are directly connected to the critical behavior of the mean-field interface position and can provide a valuable insight on the nature of surface interactions and critical wetting phenomena in nematic liquid crystals. Possible changes in the local director mode properties resulting from the critical interface position fluctuations and order electricity effects are also discussed.

PACS: 61.30.Cz, 64.70.Md

I. INTRODUCTION

Liquid crystals in confined geometries (such as plates, droplets, porous glasses, and polymer networks) constitute an interesting class of thermodynamic systems both from experimental and theoretical viewpoints [1–4]. In particular, the generic critical wetting phenomena [5,6], studied in a large number of scalar order-parameter systems, are expected to acquire some new interesting feartures near the bulk nematic-isotropic coexistence temperature T_{NI} arising from the complicated nematic order parameter (a symmetric traceless second-rank tensor). In confined nematic liquid crystals the critical wetting transition usually appears as a *continuous interface delocalization transition* at bulk coexistence, when the nematic-isotropic interface critically intervenes into the bulk as T_{NI} is approached. Typical features of a critical interface delocalization transition are the diverging interface position and the existence of a *soft fluctuation mode* directly connected to the interface position fluctuations (for a review see, e.g., Ref. [7]). At a mean-field level and in a uniform-director approximation, the macroscopic Landau-de Gennes approach [8] reproduces the well-known critical wetting results originally established for scalar order-parameter systems, so that many known results can be directly applied to nematic liquid crystals (for a review on wetting transitions in liquid crystals see Ref. [9] and references therein).

Concerning the fluctuation dynamics, the situation in systems with an orientational rong-range order might be substantionally changed. Generally, the nematic order-parameter fluctuations are characterized by five independent fluctuation fields, one of them being related to the scalar order-parameter fluctuations, whereas the other four fields describe the transverse fluctuations of the nematic director. Therefore, an interesting issue is to examine nematic director fluctuations in a regime of critical wetting, as one can expect that some peculiarities of normal-mode director fluctuations will reflect the criticalities. Published experimental works show quite different elementary excitation spectra, ranging from very small to very large relaxation rates as compared to bulk excitation spectra [10,11]. Up to now, relatively small number of theoretical studies treating the fluctuation dymamics in confined liquid crystals near the bulk coexistence have been published. Recently, Ziherl and Žumer [12] have published a work dealing with order-parameter pretransitional fluctuations above T_{NI} under the condition of a strong homeotropic anchoring of the nematic director. Apart from the typical soft fluctuation mode, reflecting the interface position fluctuations, their numerical analysis also indicated slow uniaxial director modes located close to the substrate. In the discussed work the critical wetting regime is ensured by a strong scalar order-parameter anchoring at the surface, describing substrates enhancing the nematic order. Another extreme type of substrate anchoring yet allowing for a critical wetting regime, called here *weak-anchoring limit*, corresponds to substrates strongly suppressing the nematic arrangement. For example, substrates treated by the S_iO-evaporation technique may produce such disorder effects. In this case (and under some additional conditions discussed below), it is the isotropic phase which wets the wall, so that one may expect different behavior of the normal fluctuation modes.

A goal of the present work is to compare the fluctuation mode dynamics based on such extremely different substrate potentials. The emphasis is especially on substrates strongly suppressing the nematic order. A characteristic feature of the weak anchoring is the possible critical behavior of the surface scalar order parameter by itself. This may also be of importance in the critical wetting experiments, as the finite size effects frequently mask the weak logarithmic divergence in the interface position (characteristic for short-ranged substrate potentials) [13]. As argued below, in the weak-anchoring limit, when the isotropic phase wets completely the substrate, in addition to the scalar order-parameter soft excitation, *gapped biaxial director modes* located in the presurface quasi-isotropic layer can exist. Basically, the low-energy spectrum of these local biaxial excitations is determined by the mean-field interface position. Since the latter is logarithmically divergent at $T = T_{NI}$, the excitation spectrum shows a characteristic cusplike behavior in that limit, as found also numerically in the strong-achoring case [12].

The cited results are based on the Gaussian approximation and suggest constant nematic-director mean-field configurations, when the director fluctuations decouple from the "dangerous" scalar order-parameter modes. In principle, a series of effects can invalidate the above two approximations. At a first place, higher-order fluctuation effects may be of importance since $D = 3$ is the upper critical spatial dimension for a critical wetting transition. In particular, it is well-known that the effective nematic-isotropic interfacial width diverges in $D = 3$ [14]. Clearly, this latter divergence can modify the local excitation spectra. However, close enough to T_{NI} the simple Gaussian picture of local director excitations is still valid, since the effective interfacial width is much smaller than the domain thickness (see below). As to the inhomogeneous director configurations, they can result from many sources, the main effect being the appearence of intermode couplings. As an example, we will discuss below order-electricity effects, as the problem is characterized by strong scalar order-parameter variations in the interfacial region.

The rest of the paper is organized as follows. In Section II we define the surface Landau-de Gennes free-energy functional and the free-energy substrate potentials describing the strong- and weak-anchoring limits. For uniform-director configurations, we present the mean-field scalar order-parameter profiles which are later used to fix the relevant areas of the phase diagrams. In Section III we study the Gaussian order-parameter fluctuations. The emphasis is on the properties of the local nematic-director excitations in the two cases of substrate anchorings and in complete wetting regimes. The role of interface position fluctuations and order electricity effects are also analyzed here. The last Section contains discussions and a summary of the results.

II. LANDAU-DE GENNES MODEL

We adopt the macroscopic Landau-de Gennes description (one elastic-constant approximation) based on the following free-energy density [8]:

$$f_{LG}(\mathbf{Q}) = \frac{L}{2}(\partial_\alpha Q_{\beta\gamma})^2 + \frac{a}{2}Q_{\alpha\beta}Q_{\alpha\beta} - \frac{b}{3}Q_{\alpha\beta}Q_{\beta\gamma}Q_{\gamma\alpha} + \frac{c}{4}(Q_{\alpha\beta}Q_{\alpha\beta})^2. \tag{1}$$

Here $\mathbf{Q} = Q_{\alpha\beta}(\mathbf{r})$ denotes the nematic order-parameter field (symmetric traceless second-rank tensor), L is an elastic constant, and $a \equiv \alpha(T - T^*)$. The material constants α, b, and c are supposed to be positive and temperature-independent. T^* is the supercooling temperature for the bulk isotropic phase.

It is convenient to introdice the following reduced quantities: (i) the reduced tensor order parameter $\mathbf{S} \equiv \mathbf{Q}/Q_c$, where $Q_c = 2b/3\sqrt{6}c$ is the value of the scalar order parameter Q at the bulk nematic-isotropic phase-transition temperature $T_{NI} = T^* + b^2/27\alpha c$, (ii) the reduced temperature $\tau = (T - T_{NI})/(T_{NI} - T^*)$. The liquid crystal is assumed to occupy the semi-infinite space $z > 0$, z being the coordinate normal to the surface. Using the above reduced quantities, the surface free-energy functional can be recasted to the following form:

$$F_s[\mathbf{S}] = LQ_c^2 \int dxdy \int_0^\infty dz \left\{ \frac{1}{2}(\partial_\alpha S_{\beta\gamma})^2 + \frac{1}{\xi_0^2}\left[f(\mathbf{S}) - f(\mathbf{S}_b)\right] + \delta(z)f_0(\mathbf{S}) \right\}, \tag{2}$$

where the uniform free-energy potential $f(\mathbf{S})$ reads

$$f(\mathbf{S}) = \frac{1+\tau}{2}\mathrm{tr}\,\mathbf{S}^2 - \sqrt{6}\mathrm{tr}\,\mathbf{S}^3 + \frac{1}{2}\left[\mathrm{tr}\,\mathbf{S}^2\right]^2. \tag{3}$$

Here $\mathbf{S}_b = \mathbf{S}_b(\tau)$ denotes the order parameter deeply in the bulk ($z \to \infty$). The correlation length of the isotropic phase at coexistence is denoted by $\xi_0 = \sqrt{L/\alpha(T_{NI} - T^*)}$. The short-ranged substrate potential $f_0(\mathbf{S})$ (defined at $z = 0$) comes from a variation of the intermolecular forces near the surface. Up to second order in $\mathbf{S}$, $f_0(\mathbf{S})$ is frequently modeled by the expression [15]

$$f_0(\mathbf{S}) = \frac{a_s}{2}\mathrm{tr}\,(\mathbf{S} - \mathbf{S}_s)^2, \tag{4}$$

where $\mathbf{S}_s$ is a symmetric traceless second-rank tensor characterizing the substrate, and a_s is a phenomenological constant related to the ratio of the coupling constants at the surface to those in the bulk.

The Euler-Lagrange equation related to Eq. (2) reads

$$-\xi_0^2 \Delta\mathbf{S} + (1+\tau)\mathbf{S} + 3\sqrt{6}\left[\mathbf{S}^2 - \frac{1}{3}\left(\mathrm{tr}\,\mathbf{S}^2\right)\mathbf{1}\right] + 2\left(\mathrm{tr}\,\mathbf{S}^2\right)\mathbf{S} = 0, \tag{5}$$

where $\mathbf{1}$ is the unit second-rank tensor. The boundary condition at $z = 0$, related to the last equation, is

$$\left[-\frac{d\mathbf{S}}{dz} + \frac{\partial f_0(\mathbf{S})}{\partial\mathbf{S}}\right]\bigg|_{z=0} = 0. \tag{6}$$

A. Uniform-director order-parameter profiles

In a uniaxial uniform-director mean-field configuration, the solution of Eq. (5) has the following generic form:

$$\mathbf{S}_0(z) = \frac{u(z)}{\sqrt{6}}(3\mathbf{n}_0 \otimes \mathbf{n}_0 - \mathbf{1}), \tag{7}$$

where $\mathbf{n}_0$ is the constant nematic director and $u(z)$ is the scalar order-parameter profile. As discussed below, the uniform-director ansatz (7) is presumably not valid when Eq. (2) contains terms imposing elastic nematic-director distorsions (such as order-electricity or flexoelectric free-energy terms). Nevertheless, the uniform-director approximation is a useful starting point even in the presence of such terms, as far as the system is in a regime of critical wetting and close to T_{NI}.

For future references, we briefly study below the mean-field solutions of Eq. (5) as well as the surface phase diagrams (for the two extreme cases of surface free-energy potentials discussed above) in a vicinity of the bulk transition temperature T_{NI}. In a uniform-director approximation, Eq. (5) reduces to the following non-linear differential equation for the profile function $u(z)$:

$$-\frac{d^2u}{dz^2} + \frac{1}{\xi_0^2}\frac{\partial f(u)}{\partial u} = 0, \tag{8}$$

$$f(u) = \frac{1+\tau}{2}u^2 - u^3 + \frac{1}{2}u^4, \tag{9}$$

supplemented by the boundary condition

$$\frac{du}{dz}\bigg|_{z=0} = \frac{\partial f_0}{\partial u_0}. \tag{10}$$

Here u_0 is the value of the scalar order parameter $u(z)$ at the surface, $u_0 \equiv u(z)_{z=0}$. Deeply in the bulk the conditions read $(du/dz)_{z=+\infty} = 0$ and $u(z = +\infty) = u_b$, where $u_b = u_b(\tau)$ is the bulk scalar order parameter: $u_b = 0$ in the isotropic phase and

$$u_b(\tau) = \frac{1}{4}\left(3 + \sqrt{1 - 8\tau}\right), \qquad \tau \le 0, \tag{11}$$

in the ordered nematic phase.

It is instructive to rewrite Eq. (8), by use of the boundary conditions for $u(z)$, in the form

$$\xi_0\frac{du}{dz} = \mathrm{sign}(u_b - u_0)\sqrt{2f(u) - 2f(u_b)}, \tag{12}$$

where $\mathrm{sign}(u_b - u_0) = \pm 1$ for $u_b > u_0$ and $u_b < u_0$, respectively. Now, using Eq. (12), the boundary condition (10) can be transformed to an implicit equation for u_0,

$$\text{sign}(u_b - u_0)\frac{1}{\xi_0}\sqrt{2f(u_0) - 2f(u_b)} = \frac{\partial f_0}{\partial u_0}, \tag{13}$$

and the free-energy functional (2) reduces to the following function of u_0:

$$f_s(u_0) \equiv \frac{F_s[u_0]}{LQ_c^2} = \frac{\text{sign}(u_b - u_0)}{\xi_0}\int_{u_0}^{u_b} du\sqrt{2f(u) - 2f(u_b)} + f_0(u_0). \tag{14}$$

In terms of the function $f_s(u_0)$, Eq. (13) is equivalent to the minimum condition $df_s(u_0)/du_0 = 0$, whereas the phase stability condition reads

$$\frac{d^2 f_s(u_0)}{du_0^2} \equiv \frac{\text{sign}(u_0 - u_b)}{\xi_0\sqrt{2f(u) - 2f(u_b)}}\frac{df(u_0)}{du_0} + \frac{d^2 f_0(u_0)}{du_0^2} > 0. \tag{15}$$

The last three equations completely describe the surface phase diagram of a semi-infinite nematic liquid crystal in uniform-director uniaxial states. In the context of surface phase transitions in nematic liquid crystals, using the substrate potential $f_0(u_0) = -h_0 u_0$, these equations were originally analyzed by Sheng [16]. Surface free-energy potentials of the form

$$f_0(u_0) = -h_0 u_0 + \frac{a_s}{2}u_0^2, \tag{16}$$

constitute the basis of the theory of *wetting fenonomena* in simple liquids [5] and they have also been used in the context of liquid crystals by Sluckin and Poniewierski [17]. Since the emphasis in the present study is on fluctuation effects in a regime of critical wetting, we do not analyze the full phase diagram related to Eq. (4). Instead, we mainly consider uniaxial surface potentials which can be described by the tensor parameter

$$\mathbf{S}_s = \frac{u_s}{\sqrt{6}}(3\mathbf{n}_s \otimes \mathbf{n}_s - \mathbf{1}) \tag{17}$$

in Eq. (4), where u_s and $\mathbf{n}_s$ are the scalar order parameter and the director orientation preffered at the surface. To be concrete, in what follows we assume that the unit vector $\mathbf{n}_s$ is normal to the surface (homeotropic director anchoring). In the cases without elastic distorsion forces, such as those described by the free-energy potential (1) under appropriate boundary conditions, the director $\mathbf{n}_0$ is always parallel to $\mathbf{n}_s$, provided that $u_s > 0$. In this case Eq. (4) is reduced to the following substrate potential:

$$f_0(u_0) = \frac{a_s}{2}(u_0 - u_s)^2, \qquad a_s \geq 0. \tag{18}$$

There are two interesting limits of Eq. (18) when the solution of Eq. (13) is extremely simple: (i) The *srong-anchoring limit*, $a_s \to \infty$, which is appropriate for substrates imposing a strong scalar order-parameter anchoring at the surface. In this case the solution of Eq. (13) just reads $u_0 = u_s$. This is also the case discussed in Ref. [12]. (ii) The second interesting limit of Eq. (18) corresponds to the choice $u_s = 0$. This latter condition may fit to, say, substrates treated by S_iO- evaporation technique, when the nematic order is strongly suppressed. In this case Eq. (4) reduces to $f_0(\mathbf{S}) = (a_s/2)\text{tr}\,\mathbf{S}^2$, so that the substrate does not impose any preferred direction on the nematic director. We call this *weak-anchoring limit*. It is important that the above simplified potentials generate the main critical wetting regimes [7] found with the more general form (16), so that we restrict our interest to the above extremely different cases.

For the bulk free-energy potential (9) and for uniform-director mean-field configurations, the non-linear differential equation (12) can be solved both above and below T_{NI} and for arbitrary u_0. The surface scalar order parameter u_0 should be independently obtained from Eqs. (13) and (18). The explicit expression for the order-parameter profile above T_{NI} reads

$$u(z) = \frac{1 + \tau}{1 + \sqrt{\tau}\,\sinh\left(z/\xi_I + \alpha_I\right)}, \qquad \tau > 0, \tag{19}$$

$$\alpha_I = \text{arcsinh}\left[\frac{1}{\sqrt{\tau}}\left(\frac{1 + \tau}{u_0} - 1\right)\right],$$

where $\xi_I = \xi_0/\sqrt{1 + \tau}$ is the correlation length in the isotropic phase.

Below the bulk coexistence temperature T_{NI} the profile function reads

$$u(z) = u_b - \frac{3u_b - 2(1+\tau)}{2u_b - 1 - \sqrt{u_b - 1}\sinh\left[\mathrm{sign}(u_0 - u_b)z/\xi_N + \alpha_N\right]}, \qquad \tau < 0, \tag{20}$$

$$\alpha_N = \mathrm{arcsinh}\left\{\frac{1}{\sqrt{u_b - 1}}\left[2u_b - 1 + \frac{3u_b - 2(1+\tau)}{u_0 - u_b}\right]\right\},$$

where $\xi_N \equiv d^2 f(u_b)/du_b^2 = \xi_0/\sqrt{3u_b - 2(1+\tau)}$ is the correlation length in the nematic phase. For $\tau > 0$ the profile $u(z)$ describes a paranematic layer, whereas the bulk is occupied by the isotropic phase. For large enough u_0, $u(z)$ has an inflection point at $z = d_I$ marking the center of the nematic-isotropic interface. The implicit equation for d_I is $u(d_I) = u_1$, where the function $u_1 = u_1(\tau) = (3 - \sqrt{1 - 8\tau})/4$ corresponds to the local maximum of $f(u)$, Eq. (9). Explicitly,

$$d_I = \xi_I \ln\left|\left(\frac{u_0}{u_1}\right)\frac{\tau + (1 - u_1) + \sqrt{(1+\tau)\left[\tau + (1 - u_1)^2\right]}}{\tau + (1 - u_0) + \sqrt{(1+\tau)\left[\tau + (1 - u_0)^2\right]}}\right|, \qquad u_0 > u_1. \tag{21}$$

Similar formulae can be established below T_{NI}. For $u_0 < u_b$ the surface layer is less ordered as compared to the bulk. If in addition $u_0 < u_1$, the profile has an inflection point at $z = d_N$ marking the interface position difined by the equation $u(d_N) = u_1$ (see Fig. 1). The result reads

$$d_N = \xi_N \ln\left|\left(\frac{u_b - u_0}{u_b - u_1}\right)\frac{u_1 u_b + (u_b - 1)(2u_b + u_1) + \sqrt{u_b(4u_b - 3)\left[u_b - 1 + (u_b + u_1 - 1)^2\right]}}{u_0 u_b + (u_b - 1)(2u_b + u_0) + \sqrt{u_b(4u_b - 3)\left[u_b - 1 + (u_b + u_0 - 1)^2\right]}}\right|. \tag{22}$$

B. Surface phase diagrams near T_{NI}

The profile functions, Eqs. (19) and (20), may be used to describe the surface phase diagrams. In the context of interface delocalization transitions [18], the surface phase diagram has already been analyzed in the framework of the substrate potential (16) [7]. For our study it is enough to consider the extreme cases specified above and to mark the respective peculiarities connected to the form of the used surface free-energy potentials.

(i) *Strong-anchoring limit*:

Firstly, one must find the surface order parameter u_0 from Eq. (13). In the limit $a_s \to +\infty$ the solution of Eq. (13) just reads $u_0 = u_s$, so that the surface is characterized by the parameter u_s. Let us consider the phase diagram for $\tau > 0$. If $u_s > 1$ and $\tau \to 0^+$, Eq. (21) predicts a logarithmically diverging interface position given by $d_I = \xi_0 \ln(1/\tau) + O(1)$. Exactly at $u_s = 1$ one finds $d_I = (\xi_0/2)\ln(1/\tau) + O(1)$. The critical behavior is connected to the following non-analytic terms in the surface free energy (14) considered as a function of τ and $\Delta \equiv u_s - 1$:

$$f_s^{sing}(\tau, \Delta) = \frac{\tau}{2}\ln\frac{\Delta + \sqrt{\Delta^2 + \tau}}{\tau} + \frac{\Delta}{2}\sqrt{\Delta^2 + \tau} + \frac{1}{3}(\Delta^2 + \tau)^{3/2}. \tag{23}$$

In the region $\Delta < 0$ there are no singular terms at all, and the thickness of the presurface nematic layer d_I is of the order of the correlation length ξ_I (partial wetting). On the other hand, one finds $f_s^{sing} \propto \tau \ln(1/\tau)$ as $\Delta \geq 0$. Thus, there is a line of critical points, defined by $\tau = 0^+$ and $u_s \geq 1$, describing a critical wetting of the substrate by the nematic phase. The transition at $\tau = 0^+$ for $u_s > 1$ is known as a *complete wetting* or as a *critical interface delocalization transition* of type P^+. [7] The point $(\tau, \Delta) = (0^+, 0)$ is a special point on the phase diagram since the parameter Δ becomes a relevant field (in addition to τ): the derivative $\partial^2 f_s/\partial\Delta^2 \propto \Delta/\sqrt{\tau + \Delta^2}$ has a finite jump at $\Delta = 0$. Respectively, the interface position $d_I = \xi_0 \ln(1/|\Delta|) + O(1)$ is finite as $\Delta < 0$, whereas it is delocalazed (infinite) when $\Delta > 0$. As a matter of fact, the multicritical point $(\tau, \Delta) = (0^+, 0)$ corresponds to the tricritical point T^+ found for the substrate potential (16), but in the strong-anchoring limit $(a_s \to \infty)$ the summetry breaking field h_0 is irrelevant.

Similar critical properties can be established just below the bulk coexistence temperature T_{NI}. Now the interface is located at $z = d_N$, and the region $0 < z < d_N$ is occupied by a suppressed (as compared to the bulk) nematic phase, provided that $u_s < u_1$. For positive values of the parameter u_s, d_N remains finite at $\tau = 0^-$, and the thickness of the surface layer d_N is of the order of ξ_N. Complete wetting of the substrate by the isotropic phase is possible only for non-positive values of u_s [19]. The point $(\tau, u_s) = (0^-, 0)$ is a multicritical point analogous to those discussed above.

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The interface position d_N shows the same asymptotic behavior as d_I does: for $u_s < 0$ and $\tau \to 0^-$, it diverges as $d_N = \xi_0 \ln(1/|\tau|) + O(1)$, whereas at $u_s = 0$ one has $d_N = (\xi_0/2)\ln(1/|\tau|) + O(1)$. For fixed $\tau = 0^-$, d_N is finite when $u_s > 0$, $d_N = \xi_0 \ln(1/u_s) + O(u_s)$, and it becomes infinite as $u_s < 0$.

(ii) *Weak-anchoring limit*:

This case is more interesting in a sense that now the surface order parameter u_0 is not pinned by the surface and may change with the temperature, $u_0 = u_0(\tau)$. As obtained from the inplicit Eq. (13), the function $u_0(\tau)$ has the following asymptotic behavior. For large enough a_s ($a_s > \xi_0^{-1}$) and $\tau \to 0^-$, $u_0 = u_0(\tau)$ vanishes as

$$u_0(\tau) = \frac{1}{\sqrt{\xi_0^2 a_s^2 - 1}}|\tau|^{\frac{1}{2}} + O(|\tau|), \quad a_s \xi_0 > 1, \quad \tau < 0, \tag{24}$$

whereas $u_0 = 0$ all over the region $\tau > 0$. This additional surface criticality is specific for the weak-anchoring limit and takes place simultaneously with the critical interface delocalization discussed above. The asymptotic behavior of d_N as $\tau \to 0^-$ (see Fig. 1) is analogous to those found in the case of srong anchoring: $d_N = \xi_0 \ln(1/u_0) + O(1)$, with $u_0 = u_0(\tau)$ from Eq. (24).

The case $a_s < \xi_0^{-1}$ is less interesting in a sense that now the surface order parameter u_0 is discontinuous at T_{NI}: it jumps from the finite value $u_0 = 1 - a_s\xi_0 + O(|\tau|)$ (for $\tau \to 0^-$) to zero (for $\tau > 0$). Respectively, there is no critical behavior for $a_s\xi_0 < 1$, and d_N is of the order of the correlation length in the nematic phase ξ_N. The point $(\tau, a_s\xi_0) = (0^-, 1)$ is a milticritical point (type T^- according to Ref. ([7])) characterized by three relevant fields, i.e., τ, $a_s\xi_0$, and h_0. Exactly at $a_s\xi_0 = 1$, the surface order parameter $u_0(\tau)$ acquires the asymptotic behavior

$$u_0(\tau) = \left|\frac{\tau}{2}\right|^{\frac{1}{3}} + O(|\tau|), \quad a_s\xi_0 = 1, \quad \tau < 0. \tag{25}$$

Respectively, the interface position delocalizes as $d_N = \xi_0 \ln(1/u_0) + O(1)$, with $u_0 = u_0(\tau)$ defined by Eq. (25).

III. LOCAL FLUCTUATION MODES

Within the Landau-de Gennes approach the normal fluctuation modes are obtained by solving the eigenvalue problem connected to the linearized Euler-Lagrange equation. The latter equation can be derived from Eq. (5) by use of the order-parameter decomposition

$$\mathbf{S}(\mathbf{r}) = \mathbf{S_0}(z) + \boldsymbol{\phi}(\mathbf{r}). \tag{26}$$

Here $\mathbf{S_0}(z)$ is the mean-field solution (7), as defined by the scalar order parameter $u(z)$, Eqs. (19) and (20). $\boldsymbol{\phi}(\mathbf{r})$ is the order-parameter fluctuation field (symmetric traceless second-rank tensor).

It is useful to adopt the following parametrization of $\boldsymbol{\phi}(\mathbf{r})$ [20]:

$$\boldsymbol{\phi}(\mathbf{r}) = \sum_{i=0}^{4} \phi_i(\mathbf{r})\mathbf{g}_i, \tag{27}$$

where the base tensors $\mathbf{g}_i$, $i = 0, \cdots, 4$ read

$$\mathbf{g}_0 = \frac{1}{\sqrt{6}}(3\mathbf{n}_0 \otimes \mathbf{n}_0 - \mathbf{1}),$$

$$\mathbf{g}_1 = \frac{1}{\sqrt{2}}(\mathbf{e}_1 \otimes \mathbf{n}_0 + \mathbf{n}_0 \otimes \mathbf{e}_1), \quad \mathbf{g}_2 = \frac{1}{\sqrt{2}}(\mathbf{e}_2 \otimes \mathbf{n}_0 + \mathbf{n}_0 \otimes \mathbf{e}_2), \tag{28}$$

$$\mathbf{g}_3 = \frac{1}{\sqrt{2}}(\mathbf{e}_1 \otimes \mathbf{e}_1 - \mathbf{e}_2 \otimes \mathbf{e}_2), \quad \mathbf{g}_4 = \frac{1}{\sqrt{2}}(\mathbf{e}_1 \otimes \mathbf{e}_2 + \mathbf{e}_2 \otimes \mathbf{e}_1),$$

and satisfy the orthogonality relation $\mathrm{Tr}\,(\mathbf{g}_i.\mathbf{g}_j) = \delta_{ij}$. The unit vectors $\mathbf{e}_1 \perp \mathbf{e}_2$ are perpendicular to the nematic director $\mathbf{n}_0$.

The variable $\phi_0(\mathbf{r})$ describes *longitudinal* scalar order-parameter fluctuations, whereas the pair variables $[\phi_1(\mathbf{r}), \phi_2(\mathbf{r})]$ and $[\phi_3(\mathbf{r}), \phi_4(\mathbf{r})]$ are connected to the *transverse* uniaxial and biaxial director fluctuations, respectively.

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Due to the symmetry in the (x, y) plane, it is suitable to work in a mixed $(\mathbf{k}_\perp, z)$ representation, i.e., $\phi_i = \phi_i(\mathbf{k}_\perp, z)$, where $\mathbf{k}_\perp = (k_x, k_y)$ is the wave vector in the (x, y) plane. Now, projecting the linearized Euler-Lagrange equation on the base tensors $\mathbf{g}_i$, one obtains the following independent Schrödinger-type eigenmode equations:

$$\left[-\xi_0^2 \frac{d^2}{dz^2} + V_i(z) \right] \phi_i(z) = E_i \phi_i(z), \quad i = 0, \cdots, 4, \tag{29}$$

where the functions $V_0(z) = 1 + \tau - 6u(z) + 6u(z)^2$, $V_1(z) = V_2(z) = 1 + \tau - 3u(z) + 2u(z)^2$, and $V_3(z) = V_4(z) = 1 + \tau + 6u(z) + 2u(z)^2$ may be thought of as potential energies in a related quantum-mechanical problem. The profile function $u(z)$ is defined by Eqs. (19) and (20). The correlation functions of the order-parameter fields $G_i(k_\perp; z, z') = \langle \phi_i(\mathbf{k}_\perp, z) \phi_i(\mathbf{k}_\perp, z') \rangle$ satisfy the equations

$$\left[k_\perp^2 - \xi_0^2 \frac{d^2}{dz^2} + V_i(z) \right] G_i(k_\perp; z, z') = \delta(z - z'), \quad i = 0, \cdots, 4, \tag{30}$$

and can be represented, using the eigenmodes $\phi_i^{(n)}(z)$ and the eigenvalues $E_i^{(n)}$ of Eq. (29), in the form

$$G_i(k_\perp; z, z') = \sum_n \frac{\phi_i^{(n)*}(z) \phi_i^{(n)}(z')}{k_\perp^2 + E_i^{(n)}}, \quad i = 0, \cdots, 4. \tag{31}$$

In what follows we study the eigenvalue problem in the two extreme cases of surface free-energy potentials specified above. In particular, we argue that the local uniaxial director excitations are characteristic of surface potentials favoring the nematic state, whereas in the case of weak-anchoring potentials suppressing the nematic order, as a rule, the local biaxial excitations are favored. In principle, the mode spectrum may be disturbed by several issues, such as higher-order fluctuation effects, finite-size corrections, long-range substrate forces, and the order electricity. Some of the mentioned effects modifying the local director excitations are discussed below. As to the external electromagnetic fileds, this problem deserves a separate study.

A. Strong-anchoring limit: local uniaxial director modes

As discussed above, for substrates enhancing the scalar order parameter ($u_0 = u_s > 1$) (and for a fixed surface homeotropic orientation of $\mathbf{n_0}$), the surface phase diagram contains a line of critical points describing a complete wetting. The related eigenvalue problem, Eq. (29), has recently been numerically analyzed by Žiherl and Žumer [12]. In this strong-anchoring case, the fluctuation modes are pinned at the surface, i.e., $\phi_i(z)|_{z=0} = 0$, $i = 1, \cdots, 4$. Two types of local exitations, related to the potential wells shown in Fig. 2, have been established. The first one is the lowest soft mode $\phi_0^{(0)}(z)$ characterized by an energy $E_0^{(0)}(\tau) \propto \tau$ and related to the scalar order-parameter field: This mode describes fluctuations of the mean-field interface position located at $z = d_I$. Physically, this excitation appears as a result of the broken translational symmetry [$u(z) \neq 0$] and its existence is a typical feature of the critical wetting transition. The second type of localized modes found in the discussed work are connected with the uniaxial director fluctuation fields $\phi_1(z)$ and $\phi_2(z)$. In the vicinity of T_{NI} the uniaxial modes soften and become gapless at T_{NI}. As seen below, these features of the local uniaxial director excitations can easily be obtained analytically in the limit $\tau \to 0^+$.

The physics behind the local uniaxial director modes is simple. At a mean-field level and in the limit $\tau \to 0^+$, the interface width, as a rule being of the order of the correlation length ξ_0, is much smaller than the layer thickness $d_I \simeq \xi_0 \ln(1/\tau)$, so that the region $0 < z < d_I$ may be thought of as a nematic plate of width d_I, Eq. (21). In the same limit, and for the low-energy excitations, one can use the following simpified form of the potential $V_1(z)$ (see Fig. 2): $V_1(z) \approx 0$ in the interval $0 < z < d_I$, and $V_1(z) \approx 1$ for $z > d_I$. Thus, using the continuity of the logarithmic derivative of the field $\phi_1(z)$ at $z = d_I$, one finds the excitation energies $E_1^{(n)} = \xi_0^2 k_n^2$. The parameter k_n satisfies the implicit equation

$$k_n d_I = \pi n - \arcsin(k_n \xi_0), \quad n = 1, 2, \cdots, n_{max}, \tag{32}$$

where the number of localized modes n_{max} is finite, fixed by the condition $0 \leq k_n \xi_0 \leq 1$, and dependent on the reduced temperature τ (since $d_I = d_I(\tau)$). In the limit $\tau \to 0^+$, when $d_I \simeq \xi_0 \ln(1/\tau)$, the excitation energies $E_1^{(n)}$ take the followind asymptotic form:

$$E_1^{(n)} \simeq \frac{\pi^2 n^2}{\ln^2(1/\tau)}, \qquad n = 1, 2, \cdots, n_{max}. \tag{33}$$

This expression is in agreement with the numerical results of Ref. [12] and reproduces, in particular, the observed cusplike bihavior of the low-laying energy levels. We see that the low-energy levels give a direct information about the logarithmically divergent interface position, $d_I \simeq \xi_0 \ln(1/\tau)$. This critical behavior is characteristic for the short-ranged substrate interactions. For example, the Van der Waals type interactions imply the power-low critical behavior $d_I \propto \tau^{-1/3}$, whereas the Fréedericksz-type wetting transition was shown to give the asymptotic form $d_I \propto \tau^{-1/2}$ [21]. It is clear that the above excitation spectra can give a valuable information on both the nature of presuface forces and the criticality by itself.

B. Weak-anchoring case: local biaxial director modes

Let us remember that in the case of weak-anchoring substrate potentials the complete wetting regime is realized for $a_s \geq \xi_0^{-1}$ as $\tau \to 0^-$. Now it is the isotropic phase which wets the wall and the presurface layer of thickness $d_N = d_N(\tau)$ may be thought of as a plate occupied by the isotropic phase. As in the limit of strong anchoring, the critical behavior is controlled by the lowest scalar order-parameter mode $\phi_0^{(0)}$ describing fluctuations of the mean-field interface position at $z = d_N$. A good approximation for $\phi_0^{(0)}$ can be obtained in the limit $\tau \to 0^-$, when the interface profile $u(z)$, Eq. (20), practically coincides with the profile of the infinite system at coexistence, $u(z) \approx u^\infty(z) = \{1 + \tanh[(z - d_N)/2\xi_0]\}$ (exlcuding a small vicinity of the substrate). Since in the infinite system the function $\phi_0^\infty(z) = du^\infty(z)/dz$ is a solution of Eq. (29) with the potential $V_0(z) = V_0[u^\infty(z)]$, it is clear that the variational ansatz

$$\phi_0^{(0)}(z) = C(\tau)e^{-z/\xi_0} + \frac{1}{4\cosh^2[(z - d_N)/2\xi_0]}, \tag{34}$$

gives a good approximation for $\phi_0^{(0)}(z)$ in the limit $\tau \to 0^-$. Here the interface position $d_N = d_N(\tau)$ is defined by Eq. (22). The first term in Eq. (34) is introduced in order to satisfy the boundary condition (10), whereas the second one is a solution of Eq. (29) for the infinite system at $T = T_{NI}$. From Eq. (10) one finds that the parameter $C = C(\tau)$ takes in the limit $\tau \to 0^-$ the asymptotic form $C(\tau) = -(a_s\xi_0 - 1)u_0(\tau)$, where $u_0(\tau) \propto |\tau|^{1/2}$ is given by Eq. (24) in the case $a_s\xi_0 > 1$. Using the above ansatz as a variational function, one can show analytically [22] that the lowest excited state $\phi_0^{(0)}(z)$ has the energy $E_0^{(0)} \propto |\tau|$ and is connected to fluctuations of the mean-field interface position at $z = d_N$. More details on the importance of the discussed soft excitation will be presented in the next Subsection.

Now, let us address the nematic-director fluctuations. Due to the fact that biaxial director fluctuations are *strongly suppressed* in the bulk nematic phase (as compared to the isotropic bulk phase), there is a well in the potential function $V_3(z)$ (see Fig. 3) in the eigenmode equations (29). The potential well is located in the presurface quasi-isotropic domain with a characteristic thickness d_N. Thus, in the weak-anchoring case *bound biaxial director modes* located in the persurface domain can appear. On the other hand, the uniaxial director modes, being gapless Goldstone modes in the bulk nematic phase, are controlled by the monotonically decreasing potential $V_1(z)$, so that they will be delocalized all over the sample. In the vicinity of T_{NI} the low-energy levels of the local biaxial modes can easily be obtained analytically by use of the same procedue applied above to the uniaxial excitations in the regime of strong anchoring. Now the potential $V_3(z)$ is simplified as $V_3(z) \approx 1$ (for $0 < z < d_N$), and $V_3(z) \approx 9$ (for $z > d_N$). The boundary condition at $z = 0$ for the biaxial field $\phi_3(z)$, as obtained from Eq. (6), reads

$$\left[\frac{d\phi_3(z)}{dz} - a_s\phi_3(z)\right]_{z=0} = 0. \tag{35}$$

Using the same arguments as in the previous case, one finds the excitation energies $E_3^{(n)} = 1 + \xi_0^2 k_n^2$, where the parameter k_n now satisfies the followig implicit equation:

$$k_n d_N = \pi n - \arcsin\left(\frac{\xi_0 k_n}{2\sqrt{2}}\right) - \arcsin\left(\frac{k_n}{\sqrt{k_n^2 + a_s^2}}\right), \qquad n = 1, 2, \cdots, n_{max}. \tag{36}$$

The number of localized modes n_{max} is fixed by the condition $0 \leq k_n\xi_0/2\sqrt{2} \leq 1$, and n_{max} also depends on the reduced temperature τ through the function $d_N = d_N(\tau)$. Using the asymptotic expression $d_N \simeq \xi_0 \ln(1/|\tau|)$, one gets the following result in the limit $\tau \to 0^-$:

$$E_3^{(n)} = 1 + \frac{\pi^2 n^2}{\ln^2(1/|\tau|)}, \qquad n = 1, 2, \cdots, n_{max}. \tag{37}$$

We see that in the limit $\tau \to 0^-$ the local biaxial modes are stongly softened as compared to the bulk biaxial fluctuations in the nematic phase: For every n, the energies $E_3^{(n)}$ change from $E_3^{(n)} = 9$ (the gap in the bulk nematic phase) to $E_3^{(n)} = 1$ (the gap in the bulk isotropic phase). As expected, the local biaxial mode spectrum is gapped at $T = T_{NI}$.

C. Role of the interface-position fluctuations

Since the upper critical spatial dimension for a critical wetting transition is $D = 3$ [23,14], higher order fluctuation effects are not excluded. Indeed, as discussed above, the singular part of the mean-field free energy in the regime of complete wetting behaves like $f_s^{sing} \propto -\tau \ln(\tau)$, Eq. (23). On the other hand, the one-loop fluctuation contribution to the free energy is related to the lowest soft mode $\phi_0^{(0)}(z)$ since the energies of the excited states of Eq. (29) are separated from $E_0^{(0)}$ with a finite gap. Using this fact, it is easy to see that the fluctuation part of the free energy is $f_s^{fl}(\tau) \propto -E_0^{(0)} \ln(E_0^{(0)}) \sim -\tau \ln(\tau)$, so that it compares to the singular part of the mean-field free energy. Remember that for uniform-director configurations the scalar order-parameter field $\phi_0(\mathbf{r})$ is decoupled from the director fields in the Gaussian approximation. Therefore, in this approximation most of the results known from the scalar theory [22] can be applied to nematic systems without changes. However, the fluctuation mode $\phi_0^{(0)}$ will effectively disturb the studied local director excitations. It is easy to see this qualitatively if remember some of the fluctuation effects connected to the soft mode $\phi_0^{(0)}(z)$, originally obtained in a context of the scalar order-parameter theory. At the first place, it can be shown that this mode produces the singularities in the correation function

$$\langle \phi_0(\mathbf{r}_\perp, z_1) \phi_0(\mathbf{0}, z_2) \rangle \sim \exp(-r_\perp/\xi_\parallel) \phi_0^{(0)}(z_1) \phi_0^{(0)}(z_2), \tag{38}$$

where $\xi_\parallel = 1/\sqrt{E_0^{(0)}} \propto 1/\sqrt{\tau}$ is the characteristic length of the scalar order-parameter correlations parallel to the surface. Thus, there are critical long-range correlations of the scalar order-parameter fluctuations parallel to the surface (capillary waves). Another criticality connected to $\phi_0^{(0)}(z)$ is the predicted divergence of the interfacial width. Denoting by $d_I(\mathbf{r}_\perp) = d_I + \zeta(\mathbf{r}_\perp)$ the local interface position, it can be shown that the characteristic interface thickness $\xi_\perp \equiv \sqrt{<\zeta^2>}$ diverges as $\tau \to 0^+$ according the asymptotic form $\xi_\perp \propto \ln \xi_\parallel \propto \sqrt{d_I}$ [14]. Thise critical increase of the effective interface width, clearly, will effectively change the potential $V_1(z)$ in Eq. (29) which controls the director excitation spectrum. On the other hand, the threatment of the local fluctuation modes used throughout the paper remains valid, since close enough to T_{NI} the effective interfacial width will be much smaller than the characteristic domain thickness.

D. Role of the order electricity

There is a series of issues which might invalidate the uniform-director appoximation suggested in the above analysis [24,25]. The order electricity [26,27] belongs to this series. Since the nematic-isotropic interface is characterized by a strong variation of the scalar order parameter, one can expect that the order-electricity effect will play an important role in the wetting phenomena. Let us concentrate on the strong-anchorig limit, when the nematic director is strongly anchored along the normal to the surface. The inhomogeneity in the profile function $u(z)$, Eq. (19), generates along the axis z the electric polarization field $P_o^z = \left(r_1 n_{0_z}^2 + r_2\right)(du/dz)$, where r_1 and r_2 are the order-electric coefficients [27]. Denoting by θ the polar angle of the director $\mathbf{n_0} = (\sin\theta, 0, \cos\theta)$, it is easy to see that $\theta = \theta(z)$ since the free-energy density term f_o, connected with P_o^z, mixes $\mathbf{n_0}$ and du/dz:

$$f_o = -\frac{1}{2} P_o^z E_o^z = \frac{2\pi}{\epsilon_{zz}(\theta)} \left(r_1 \cos^2\theta + r_2\right)^2 \left(\frac{du}{dz}\right)^2. \tag{39}$$

Here $\epsilon_{zz}(\theta) = \epsilon_\perp + (\epsilon_\parallel - \epsilon_\perp)\cos^2\theta$ is the zz component of the dielectric tensor and E_o^z is the z component of the induced electic field. Combaining the last equation with the elastic term in Eq. (2), one finds the result that the elastic constant L in Eq. (2) is effectively renormalized as $L \to L_{eff}(\theta) = L + [4\pi/\epsilon_{zz}(\theta)]\left(r_1 \cos^2\theta + r_2\right)^2$. To find the function $\theta(z)$ we will follow the arguments of Ref. [27], applicable in our case for $\tau \to 0^+$. The Euler-Lagrange equation for the polar angle θ, as obtained from Eq. (2), reads

$$\frac{d^2\theta}{dz^2} = \frac{1}{6Lu^2}\frac{dL_{eff}}{d\theta}\left(\frac{du}{dz}\right)^2. \tag{40}$$

As shown above, $du/dz \propto \phi_0^\infty(z)$ is a δ-like function centered at $z = d_I$, Eq. (34). Thus, the function $\theta(z)$ can be approximated with a linear function in the region $0 < z < d_I$, excluding the vicinity of the interface and, possibly, of the surface. Since the interfacial energy is proportional to $L_{eff}^{1/2}$, the minimum condition for this energy $dL_{eff}/d\theta = 0$ also defines the average polar angle at the interface θ_0 [28]. We do not consider here possible deviations from the linear behavior of $\theta(z)$ close to the substrate [29]. Therefore, as $\tau \to 0^+$ the inhomogeneous director state is characterized by the polar angle

$$\theta(z) = \theta_0\frac{z}{d_I} \simeq \theta_0\frac{z}{\xi_0}\frac{1}{\ln(1/\tau)}, \quad \tau \to 0^+, \quad z \le d_I. \tag{41}$$

The inhomogeneous director state described above can generate several changes in the fluctuation mode dynamics. Since the local base tensors are now coordinate-dependent, $\mathbf{g}_i = \mathbf{g}_i(z)$, the normal mode equations (29) are coupled. In particular, there is a coupling between the longitudinal scalar order-parameter field $\phi_0(z)$ and the transverse director fluctuations $\phi_2(z)$ and $\phi_3(z)$. The mode coupling is, however, asymptotically small, as it is controlled by the small parameter $\xi_0/d_I \propto \ln^{-1}(1/\tau)$. As the interaction decreases logarithmically , the effect of order-electicity mode coupling can play an important role in real experiments.

IV. SUMMARY OF THE RESULTS AND DISCUSSION

In this article the emphasis was on specific features of the presurface fluctuation mode dynamics in nematic liquid crystals, arising in a regime of critical wetting when the system is close to the nematic-isotropic phase transition temperature. The importance of substratate anchoring for the director mode dynamics is demonstrated by use of two extreme types of surface free-energy potentials.

Our key results are as follows: In the case of uniform-director mean-field configurations and for a homeotropic director anchoring, surface free-energy potentials strongly suppressing the orientational order favor the appeerence of bound biaxial nematic-director fluctuation modes located in the domain occupied by the wetting thermodynamic phase. Instead, substrate forces which enhance the orientational order promote bound uniaxial modes in the presurface domain. Close to the phase coexistence temperature both types of local director excitations are strongly softened (as compared to their bulk counterparts) and acquire characteristic cusplike low-energy spectra which are directly connected to the critical behavior of the mean-field interface position. Since the physics of these local excitations closely reflects the wetting critical behavior, they can be used as a tool to examine the critical wetting phenomena and the presurface interactions in nematic liquid crystals.

We have considered one of the possible reasons for the experimentally observed different spectra of elementary exitations, ranging from very small to very large (as compared to the bulk) relaxation rates. Clearly, there could be a number of other factors modifying substantially the director exitation spectra. We have already briefly discussed two of them, i.e., the higher-order fluctuation effects and order electricity effects. As to the interface position fluctuations, in the upper spatial critical dimension $D = 3$ they lead to a divergence of the effective interface width. Thus, the local excitation specta can be disturbed, as the potentials $V_i(z)$ will be effectively changed as well. In addition, the fluctuation effects in $D = 3$ are known [14] to depend on the value of the dimensionless parameter $\omega = T/4\pi\xi\sigma$, where ξ is the bulk correlation length of the phase attracted to the wall and σ is the surface tension of the free interface. It will be interesting to see the role of this parameter in different liquid crystal materials. As to the order-electricity effects, they are expected to play an important role in the real experiments due to the strong variation of the scalar order parameter in the interfacial region. The coupling of the normal Gaussian modes is the main effect of the inhomogeneity. It is important that in this case the director fluctuation modes are coupled with the "dangerous" scalar order-parameter soft mode, so that the picture of the Gaussian fluctuation dynamics can be importantly disturbed.

In conclusion, the finite-size effects, the long-ranged substrate interactions, and the external electromagnetic fields belong to the class of factors that will modify the discussed local fluctuation mode dynamics. Clearly, these issues deserve future studies.

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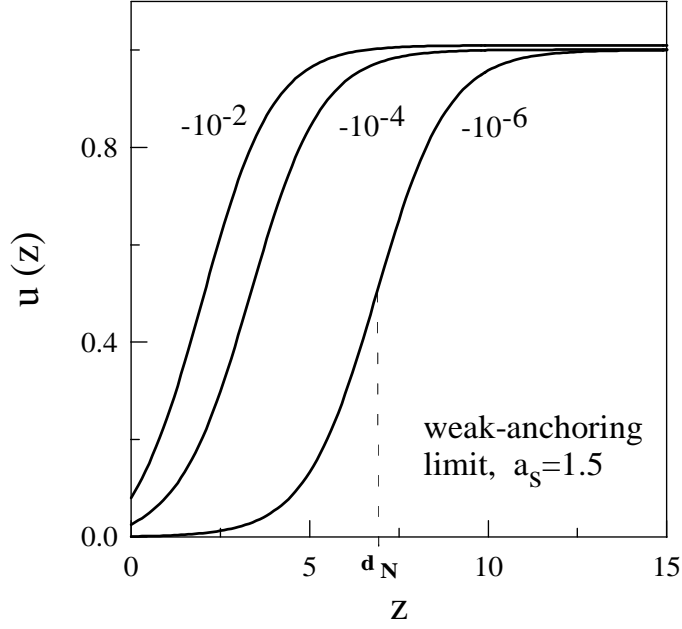[29] G. Barbero and G. Durand, J. Phys. (Paris) **47**, 2199 (1986).

FIG. 1. Scalar order-parameter profiles for the reduced temperatures $\tau = -10^{-2}$, -10^{-4}, and -10^{-6} in the case of substrates strongly suppressing the nematic order. The inflaction point at $z = d_N$ marks the characteristic isotropic domain thickness. $\xi_0 \equiv 1$
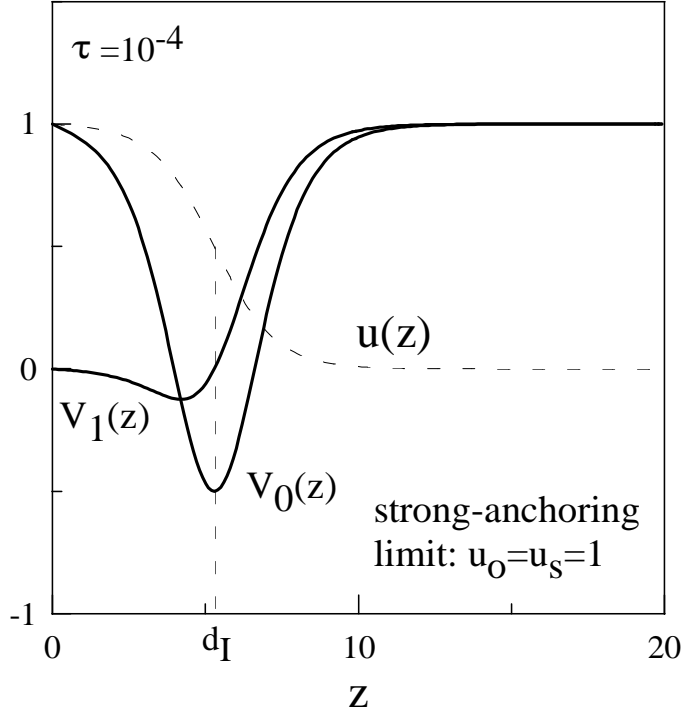
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Fig. 1
N. B. Ivanov

FIG. 2. The fluctuation mode potentials in Eq. (29) in the srtong-anchoring limit. The wells in $V_0(z)$ and $V_1(z)$ are connected with the scalar order-parameter soft mode $\phi_0^{(0)}(z)$ and the bound uniaxial director exciations $\phi_{1,2}^{(n)}(z)$, respectively. $\xi_0 \equiv 1$
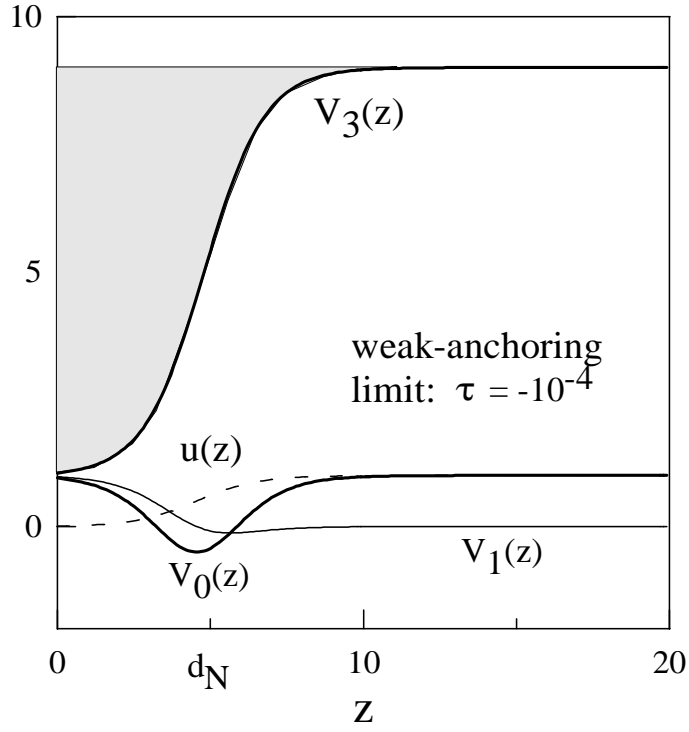
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Fig. 2
N. B. Ivanov

FIG. 3. The fluctuation mode potentials in Eq. (29) in the weak-anchoring limit. The wells in $V_0(z)$ and $V_3(z)$ (the shaded area) are connected with the scalar order-parameter soft mode $\phi_0^{(0)}(z)$ and the bound biaxial director exciations $\phi_{3,4}^{(n)}(z)$, respectively. $\xi_0 \equiv 1$

.

Fig. 3
N. B. Ivanov